LAW OFFICES
SUGHRUE, MION, ZINN, MACPEAK & SEAS, PLLC
2100 PENNSYLVANIA AVENUE, N.W.
WASHINGTON, DC 20037-3213
TELEPHONE (202) 293-7060
FACSIMILE (202) 293-7860
www.sughrue.com

October 30, 2000

BOX PCT

Assistant Commissioner for Patents
Washington, D.C. 20231

PCT/JP99/02183
-filed April 23, 1999

Re: Application of Nobuhiko TSUDA, Takahiro KITAHARA, Ryuji IWAKIRI (Deceased), Masaru NAGATO and Ryoichi FUKUGAWA
FLUORINE-CONTAINING COPOLYMER HAVING FUNCTIONAL GROUP
Our Ref: Q61520

Dear Sir:

The following documents and fees are submitted herewith in connection with the above application for the purpose of entering the National stage under 35 U.S.C. § 371 and in accordance with Chapter II of the Patent Cooperation Treaty:

☑ an English translation of the International Application.

☑ a Form PTO-1449 listing the ISR references, and a complete copy of each reference.

☑ International Search Report.

☑ International Preliminary Examination Report (foreign language).

☑ Notification Concerning Submission or Transmittal of Priority Document.

The Declaration and Power of Attorney and Assignment will be submitted at a later date.

It is assumed that copies of the International Application, the International Search Report, the International Preliminary Examination Report, and any Articles 19 and 34 amendments as required by § 371(c) will be supplied directly by the International Bureau, but if further copies are needed, the undersigned can easily provide them upon request.

The Government filing fee is calculated as follows:

Total claims	4	- 20	=		x $18.00	=	$.00
Independent claims	1	- 3	=		x $80.00	=	$.00
Base Fee							$860.00
TOTAL FEE							$860.00

A check for the statutory filing fee of $860.00 is attached. You are also directed and authorized to charge or credit any difference or overpayment to said Account. The Commissioner is hereby authorized to charge any fees under 37 C.F.R. §§ 1.16, 1.17 and 1.492

which may be required during the entire pendency of the application to Deposit Account No. 19-4880. A duplicate copy of this transmittal letter is attached.

Priority is claimed from May 1, 1998 based on Japanese Patent Application No. 122384/1998.

Respectfully submitted,

Abraham J. Rosner
Registration No. 33,276

SUGHRUE, MION, ZINN,
MACPEAK & SEAS, PLLC
2100 Pennsylvania Avenue, N.W.
Washington, D.C. 20037-3213
Telephone: (202) 293-7060
Facsimile: (202) 293-7860

Date: October 30, 2000

DESCRIPTION

FLUORINE-CONTAINING COPOLYMER HAVING FUNCTIONAL GROUP

TECHNICAL FIELD

The present invention relates to a fluorine-containing copolymer having functional group. Further the present invention relates to a fluorine-containing copolymer which is excellent in chemical resistance, solvent resistance, water resistance, weather resistance, stain-proofing property, adhesion, and the like and is suitably used for a laminating film, co-extruded layered tube, etc.

BACKGROUND ART

Fluorine-containing copolymers comprising a vinyl monomer unit having hydrocarbon type functional group are described in JP-A-1-103670, JP-A-8-41131, etc. and it is known that they are useful as crosslinking coating resins when used in combination with a curing agent. Also a fluorine-containing copolymer prepared by copolymerizing perfluorobutenoic acid or a vinyl ether having a fluorine-based functional group is known as a fluorine-containing copolymer comprising a fluorine-containing vinyl monomer.

However a fluorine-containing resin copolymer which comprises a vinyl monomer unit having hydrocarbon type functional group and is insoluble in tetrahydrofuran (THF) substantially has not been known. In case of a resin being soluble in THF, in order to realize solvent resistance, a combination use of a curing agent capable of

forming a crosslinked structure has been essential. On the other hand, from the aspect of use as a molding material, when using the copolymer together with a curing agent, it was necessary to control molding and curing reaction in a very narrow temperature range, and thus heat-molding was very difficult substantially.

Also since a monomer having a fluorine-based functional group has a good reactivity with fluoroolefin such as tetrafluoroethylene (TFE), it can introduce a functional group into a melt-moldable resin insoluble in THF such as ethylene-tetrafluoroethylene copolymer (ETFE), tetrafluoroethylene/hexafluoropropylene copolymer (FEP) and tetrafluoroethylene/perfluoro(vinyl ether) copolymer (PFA). However a preparation process of the monomer ranges over multiple stages and is disadvantageous economically.

The present inventors have found that a hydrocarbon vinyl compound monomer having functional group, particularly a vinyl ether monomer having functional group reacts with a copolymer containing a specific amount of TFE unit and can give a resin insoluble in THF, and thus completed the present invention.

DISCLOSURE OF INVENTION

Namely the present invention relates to the fluorine-containing copolymer having functional group which comprises a hydrocarbon type vinyl unit having functional group and a TFE unit and is characterized in that the fluorine-containing copolymer has a fluorine content of not less than 10 % by weight and is not dissolved in TFE substantially.

Examples of the preferred resin comprising the hydrocarbon

vinyl compound unit having functional group and TFE unit are tetrafluoroethylene (TFE), hexafluoropropylene (HFP), ethylene (ET) and hydrocarbon type vinyl ether monomer having functional group and being copolymerizable therewith.

Examples of the preferred hydrocarbon vinyl compound having functional group are functional group-containing vinyl ethers, particularly monomers having hydroxyl or epoxy.

BEST MODE FOR CARRYING OUT THE INVENTION

The fluorine-containing copolymer of the present invention is explained below.

The fluorine-containing copolymer of the present invention is a copolymer having the following characteristics (1) to (4).

(1) having TFE unit

Since the copolymer has a TFE unit, a resin having excellent chemical resistance, solvent resistance, water resistance, weather resistance and stain-proofing property can be obtained. It is preferable that the TFE unit is contained in the copolymer in an amount of not less than 5 % by mole, particularly 10 to 95 % by mole.

(2) having a fluorine content of not less than 10 % by weight

The fluorine content is derived from TFE and/or other fluorine-containing monomer. For the same reasons as in above (1), it is preferable that the fluorine content is not less than 10 % by weight, preferably 20 to 75 % by weight.

Examples of the other fluorine-containing monomer are, for instance, hexafluoropropylene, vinylidene fluoride, trifluoroethylene, monofluoroethylene, chlorotrifluoroethylene, and the like. Those

fluorine-containing monomers are optional units, and a content thereof is not more than 30 % by mole, usually 0 to 10 % by mole.

(3) being insoluble in THF substantially

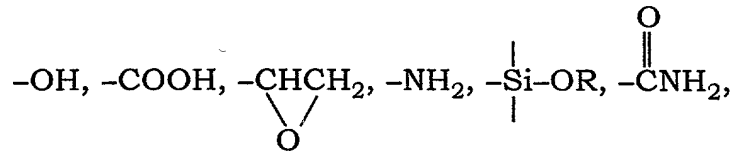
Being insoluble in THF means that the copolymer is excellent in solvent resistance. In case of a known resin being soluble in THF, in order to realize solvent resistance, a combination use of a curing agent capable of forming a crosslinked structure has been essential. On the other hand, from the aspect of use as a molding material, when using the copolymer together with a curing agent, it was necessary to control molding and curing reaction in a very narrow temperature range, and heat-molding was very difficult substantially.

Being insoluble substantially encompasses the case of being soluble in THF in a concentration of less than 0.5 % by weight. This is because when substantially measuring an intrinsic viscosity [η] of the resin in THF solution, a reliable intrinsic viscosity [η] cannot be measured unless the copolymer has a solubility of about 0.5 % by weight or more.

(4) having a hydrocarbon vinyl compound unit having functional group, preferably a hydrocarbon type vinyl ether unit having functional group

The functional group has functions of not only giving a curing site to the fluorine-containing copolymer and providing a thermoset film by reacting with a curing agent but also enhancing adhesion to various substrates. Further by laminating or co-extruding together with a resin having a site being reactive with the functional group, interface adhesion is enhanced.

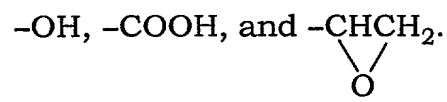
In the present invention, examples of the functional group are, for instance, functional groups represented by the formulae (i):



$-SO_3H$, and $-OSO_3H$,

wherein R is an alkyl group having 1 to 3 carbon atoms.

Among the above-mentioned functional groups, preferred are those represented by the formulae (ii):



In the present invention, examples of the hydrocarbon type vinyl ether unit having functional group are, for instance, hydroxyl-containing vinyl ether, epoxy (glycidyl)-containing vinyl ether unit, and the like.

Examples of the monomer being capable of introducing such a functional group are hydroxyl-containing monomers such as hydroxybutyl vinyl ether (HBVE) and ally alcohol; epoxy (glycidyl)-containing monomers such as glycidyl vinyl ether (GVE), and the like.

Among them, from the viewpoint of reactivity with fluoroolefin, preferred are hydrocarbon vinyl ether monomers having functional group such as HBVE and GVE.

An amount of the functional group varies depending on reactivity of the functional group and kind of a curing agent from the viewpoint of curability and adhesion when using the curing agent. It is preferable that based on the whole fluorine-containing copolymer, an

acid value is from 1 to 300 mgKOH/g, a hydroxyl value is from 1 to 200 mgKOH/g or an equivalent of epoxy is from 5 to 15,000 equivalents.

From the above-mentioned point of view, an amount of the unit giving the functional group may be selected in the range of from 0.1 to 30 % by mole, particularly 1 to 20 % by mole based on the fluorine-containing copolymer.

The fluorine-containing copolymer having functional group can be obtained by copolymerizing each of the above-mentioned monomers. The polymerization may be carried out by usual polymerization methods such as emulsion polymerization, suspension polymerization and solution polymerization. Also a monomer having functional group may be grafted on the copolymer.

Further it is preferable that the resin of the present invention has a melting point in the range of not more than 160°C when measured with DSC. If the melting point exceeds 160°C, for example, when the resin is used for a powder coating and is baked, flowability is lowered and poor appearance of a coating film such as orange peel arises.

In the case of the application of the resin of the present invention to a film, it is preferable that MFR thereof at 230°C at a load of 2.1 kg is in the range of 1 to 100 g/10 min.

Also in the case of use for lining of pipes and co-extrusion with an engineering plastic, it is preferable that MFR thereof at 160°C at a load of 2.1 kg is in the range of 1 to 100 g/10 min.

Non-restricted examples of the fluorine-containing copolymer having functional group of the present invention are those having the following combinations of monomers and satisfying the above-mentioned requirements (1) to (4).

(I) ① Perfluoroolefin not less than 5 % by mole

② Other fluorine-containing monomer 0 to 95 % by mole

③ Other non-fluorine-containing monomer 0 to 95 % by mole

④ Functional group-containing hydrocarbon type vinyl ether monomer not less than 0.1 % by mole

(II) ① At least one of TFE and HFP not less than 5 % by mole

② Other fluorine-containing monomer 0 to 95 % by mole

③ Non-fluorine-containing olefin 0 to 95 % by mole

④ Hydrocarbon type vinyl ether monomer containing functional group represented by the above-mentioned formula (i) not less than 0.1 % by mole

(III) ① At least one of TFE and HFP not less than 10 % by mole

② Other fluorine-containing monomer 0 to 20 % by mole

③ Non-fluorine-containing olefin 10 to 70 % by mole

④ Hydrocarbon type vinyl ether monomer containing functional group represented by the above-mentioned formula (i) not less than 0.1 % by mole

(IV) ① At least one of TFE and HFP not less than 20 % by mole

② Other fluorine-containing monomer 0 to 5 % by mole

③ Ethylene (ET) 20 to 50 % by mole

④ Hydrocarbon type vinyl ether monomer containing functional group represented by the above-mentioned formula (ii) not less than 0.1 % by mole

Non-restricted examples of more concrete copolymer are as follows.

TFE/HFP/ET/HBVE copolymer (mole ratio: 25 to 40/10 to 20/35 to 48/0.1 to 10) and TFE/HFP/ET/GVE copolymer (mole ratio:

25 to 40/10 to 20/35 to 48/0.1 to 10).

In the present invention, the copolymer can be used together with a curing agent which is reactable with the functional group. A hardness and resistance against stress deformation which are obtained by crosslinking can be improved.

Examples of the usable curing agent are, for instance, epoxy or glycidyl compounds such as alicyclic epoxy resin, GMA acryl, aliphatic oxysilane, triglycidyl isocyanurate (TGIC), diglycidyl terephthalate, diglycidyl p-hydroxybenzoate, spiroglycol diglycidylether and hydantoin compounds; isophorone diisocyanate, tolylene diisocyanate, xylilene diisocyanate, 4,4'-diphenylmethane diisocyanate, hexamethylene diisocyanate, dimers thereof and blocked isocyanates obtained by blocking an isocyanate group of alcohol-modified polyisocyanate with a blocking agent (for example, ε-caprolactam, phenol, benzyl alcohol, methyl ethyl ketone oxime, etc.); polybasic acid curing agent such as β-hydroxyalkylamide; polycarboxylic acids, e.g. aliphatic dibasic acids such as fumaric acid, succinic acid, adipic acid, azelaic acid, sebacic acid and dodecanedioic acid (DDA) and acid anhydrides such as phthalic anhydride, trimellitic anhydride and pyromellitic anhydride; tetramethoxymethylglycoluryl, isocyanate-modified silane coupling agent, and other curing agents described in JP-B-6-104792, JP-A-7-188587 and JP-A-1-103670.

Among them, particularly from the viewpoint of compatibility with a crosslinkable functional group in the fluorine-containing copolymer having functional group, preferred combination are as follows.

(1) Functional group: Hydroxyl group

Curing agent: Blocked isocyanate, polyurethodione

(2) Functional group: Carboxyl group

Curing agent: Triglycidyl isocyanurate, β-hydroxyalkylamide, GMA acryl

(3) Functional group: Glycidyl group

Curing agent: Aliphatic dibasic acid

It is preferable that an amount of the curing agent is from 0.1 to 1.2 equivalents, especially 0.5 to 1.0 equivalent to an amount of functional group contained in the fluorine-containing copolymer.

In addition to the curing agent, a curing catalyst may be blended. Examples of the curing catalyst are, for instance, quaternary ammonium salts such as tetrabutylammonium chloride, tetrabutylammonium bromide and tetrabutylammonium iodide; quaternary phosphonium salts such as ethyltriphenylphosphonium acetate; phosphines such as triphenylphosphine; imidazoles such as 2-methylimidazole; organotin compounds such as dibutyltindilaurate and stannous octanoate; methyltolylsulfoneimide and stannous methanesulfonate, and the like. The curing catalyst may be blended in an amount of from about 0.1 part to about 3 parts to 100 parts of the fluorine-containing copolymer having functional group.

The fluorine-containing copolymer of the present invention is also excellent in adhesion to a substrate. Examples the substrate are, for instance, various metal plates such as stainless steel plate, aluminum plate, steel plate and galvanized steel plate, and in addition, heat resistant engineering plastics such as polycarbonate, polyphenylene oxide, polyethylene terephthalate, polyether sulfone, polyamideimide and polyether ether ketone.

The fluorine-containing copolymer of the present invention is

used for various applications in the form of film. Non-restricted examples of the application are construction and building material, electric communication device, vehicles, road materials, water and gas service materials, metal products, domestic appliances, machines, tools, measuring instruments, medical instruments, utensils for maintenance, agricultural materials, ships, sports and leisure products, and the like.

Then the present invention is explained by means of examples, but is not limited to them.

EXAMPLE 1

A 4-liter pressure resistant reactor equipped with a stirrer was charged with 1,000 ml of deionized water and 1 g of potassium carbonate, and feeding of pressurized nitrogen and deairing were repeated to remove dissolved oxygen. Then the reactor was charged with 550 g of chlorofluoroethane (HCFC-141b), 7.2 g of hydroxybutyl vinyl ether (HBVE) and 650 g of hexafluoropropylene (HFP) successively. A pressure inside the reactor was increased up to 12 kgf/cm^2 at 35°C with a monomer mixture of tetrafluoroethylene (TFE)/ethylene (ET) in a % by mole ratio of 82/18. Then the reactor was charged with 1 g of cyclohexane and 48 g of 25 % flon 225 solution of isobutyryl peroxide, and a monomer mixture of tetrafluoroethylene (TFE)/ethylene (ET)/hexafluoropropylene (HFP) in a % by mole ratio of 37/43/20 was supplied continuously so that the inside pressure became constant at 12 kgf/cm^2. Every three hours after starting of the reaction, 8 g of 25 % flon 225 solution of isobutyryl peroxide was added three times. After 12-hour reaction, the inside of the reactor was restored to normal temperature and normal pressure to terminate the reaction. After the

obtained solid was washed and dehydrated, it was vacuum-dried at 80°C to give 183 g of TFE/HFP/ET/HBVE copolymer (white powder). Polymer components, melting point (Tm), MFR and solubility in THF of the obtained fluorine-containing copolymer were measured by the methods mentioned below.

Melting point: A heat balance of 10 mg of VdF polymer was measured at a heat-up speed of 10°C/min in a temperature range of -25°C to 200°C by using Thermal Analysis System (available from Perkin Elmer Co., Ltd.), and a top peak was assumed to be a melting point.

MFR: Measurement was carried out under the conditions of 160°C, a load of 2.1 kg and 10 minutes and the conditions of 230°C, a load of 2.1 kg and 10 minutes.

Solubility in THF: 0.5 Gram of resin in the form of powder was put in 10 ml of THF at room temperature, and after allowed to stand for 72 hours, dissolving state was observed with naked eyes.

The results are shown in Table 1.

EXAMPLES 2 to 3

Polymerization was carried out in the same manner as in Example 1 except that monomer components, amount of cyclohexane and polymerization time were changed as shown in Table 1. The same measurements as in Example 1 were carried out with respect to the obtained fluorine-containing copolymer. The results are shown in Table 1.

TABLE 1

Fluorine-containing copolymer having functional group	Example 1	Example 2	Example 3
Monomer components			
TFE/ET (% by mole ratio)	82/18	82/18	82/18
HFP (g)	794	794	794
HBVE (g)	21	–	21
GVE (g)	–	21	–
Amount of cyclohexane (g)	1	1	0.5
Polymerization time (hr)	12	12	7
Obtained weight (g)	183	198	167
Polymer components (% by mole)			
TFE	31	34	33
HFP	21	17	17
ET	48	44	45
HBVE	3	–	4
GVE	–	5	–
Melting point (°C)	153	157	158
MFR (g/10 min) at 230°C	77	65	96
(g/10 min) at 160°C	14	8	39
Solubility in THF	Insoluble	Insoluble	Insoluble

INDUSTRIAL APPLICABILITY

The fluorine-containing copolymer having functional group of the present invention is excellent in adhesion to a substrate and co-extrusion property, and crosslinking can be easily carried out.

CLAIMS

1. A fluorine-containing copolymer having functional group, characterized in that the copolymer contains a hydrocarbon vinyl compound unit having functional group and tetrafluoroethylene unit, has a fluorine content of not less than 10 % by weight and is insoluble in tetrahydrofuran substantially.

2. The fluorine-containing copolymer having functional group of Claim 1, wherein a melting point of the copolymer is not more than 160°C.

3. The fluorine-containing copolymer having functional group of Claim 1, wherein the copolymer contains tetrafluoroethylene unit, hexafluoropropylene unit and ethylene unit as essential components.

4. The fluorine-containing copolymer having functional group of Claim 1, wherein the hydrocarbon vinyl compound unit is hydroxybutyl vinyl ether unit or glycidyl vinyl ether unit.

Declaration and Power of Attorney for Patent Application

特許出願宣言書及び委任状

Japanese Language Declaration

日本語宣言書

下記の氏名の発明者として、私は以下の通り宣言します。

As a below named inventor, I hereby declare that:

私の住所、私書箱、国籍は下記の私の氏名の後に記載された通りです。

My residence, post office address and citizenship are as stated next to my name.

下記の名称の発明に関して請求範囲に記載され、特許出願している発明内容について、私が最初かつ唯一の発明者（下記の氏名が一つの場合）もしくは最初かつ共同発明者であると（下記の名称が複数の場合）信じています。

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

FLUORINE-CONTAINING COPOLYMER
HAVING FUNCTIONAL GROUP

上記発明の明細書（下記の欄でx印がついていない場合は、本書に添付）は、

the specification of which is attached hereto unless the following box is checked:

☐ __月__日に提出され、米国出願番号または特許協定条約国際出願番号を_____とし、
（該当する場合）_____ に訂正されました。

☑ was filed on October 30, 2000
as United States Application Number or PCT International Application Number 09/674,249
and was amended on
(if applicable)

私は、特許請求範囲を含む上記訂正後の明細書を検討し、内容を理解していることをここに表明します。

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

私は、連邦規則法典第３７編第１条５６項に定義されるとおり、特許資格の有無について重要な情報を開示する義務があることを認めます。

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56.

Declaration and Power of Attorney for Patent Application

特許出願宣言書及び委任状

Japanese Language Declaration

日本語宣言書

私は、以下に記名された発明者として、ここに下記の通り宣言する。

As a below named inventor, I hereby declare that:

私の住所、郵便の宛先及び国籍は、私の氏名の後に記載された通りである。

My residence, post office address and citizenship are as stated next to my name,

下記の名称の発明について、特許請求範囲に記載され、且つ特許が求められている発明主題に関して、私は、最初、最先且つ唯一の発明者である(唯一の氏名が記載されている場合)か、或いは最初、最先且つ共同発明者である(複数の氏名が記載されている場合)と信じている。

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

FLUORINE-CONTAINING COPOLYMER HAVING FUNCTIONAL GROUP

上記発明の明細書はここに添付されているが、下記の欄がX印でチェックされている場合は、この限りでない:

the specification of which is attached hereto unless the following box is checked:

☐ ____月____日に出願され、PCT国際出願番号を
_____ とし、かつ ____月____日に出願された
米国出願番号は_____ であり、
_____月_____日に補正された出願(該当する場合)

☒ was filed on April 23, 1999 as PCT International Application No. PCT/JP99/02183 and subsequently on October 30, 2000 as United States Application No. 09/674,249 and was amended on _____ (if applicable).

私は、上記の補正書によって補正された特許請求範囲を含む上記明細書を検討し、且つ内容を理解していることをここに表明する。

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

私は、連邦規則法典第37編1.56に定義されている特許性について重要な情報を開示する義務があることを認める。

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56.

Japanese Language Declaration

(日本語宣言書)

私は、ここに、下記に記載した外国での特許出願または発明者証の出願、或いは米国以外の少なくとも一カ国を指定している米国法典第35編365条(a)項によるPCT国際出願について、同第119条(a)-(d)項又は第365条(b)項に基づいて優先権を主張するとともに、優先権を主張する本出願の出願日よりも前の出願日を有する外国での特許出願または発明者証の出願、或いはPCT国際出願については、いかなる出願も、下記の枠内をチェックすることにより示した。

I hereby claim foreign priority under Title 35, United States Code, Section 119(a)-(d) or 365(b) of any foreign application(s) for patent or inventor's certificate, or 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Applications
外国での先行出願

Priority Not Claimed
優先権主張なし

(Number) (番号)	(Country) (国名)	(Day/Month/Year Filed) (出願日/月/年)	
122384/1998	JAPAN	1 May 1998	☐
			☐
			☐

私は、ここに、下記のいかなる米国仮特許出願についても、その米国法典第35編119条(e)項の利益を主張する。

I hereby claim the benefit under Title 35, United States Code, Section 119(e) of any United States provisional application(s) listed below.

(Application No.) (出願番号)	(Filing Date) (出願日)

私は、ここに、下記のいかなる米国出願についても、その米国法典第35編第120条に基づく利益を主張し、また米国を指定するいかなるPCT国際出願についても、その同第365条(c)に基づく利益を主張する。また、本出願の各特許請求の範囲の主題が、米国法典第35編第112条第1段に規定された態様で、先行する米国出願またはPCT国際出願に開示されていない場合においては、その先行出願の出願日と本国内出願日またはPCT国際出願日との間の期間中に入手された情報で、連邦規則法典第37編規則1.56に定義された特許性に関わる重要な情報について開示義務があることを承認する。

I hereby claim the benefit of Title 35, United States Code Section 120 of any United States application(s), or 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of Title 35, United States Code Section 112, I acknowledge the duty to disclose any material information which is material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application:

(Application No.) (出願番号)	(Filing Date) (出願日)	(Status: Patented, Pending, Abandoned) (現況: 特許許可済、係属中、放棄済)

私は、ここに表明された私自身の知識にかかわる陳述が真実であり、且つ情報と信ずることに基づく陳述が、真実であると信じられることを宣言し、さらに、故意に虚偽の陳述などを行った場合は、米国法典第18編第1001条に基づき、罰金または拘禁、もしくはその両方により処罰され、またそのような故意による虚偽の陳述は、本出願またはそれに対して発行されるいかなる特許も、その有効性に問題が生ずることを理解した上で、陳述が行われたことを、ここに宣言する。

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Japanese Language Declaration

(日本語宣言書)

委任状：私は、本出願を審査する手続きを行い、且つ米国特許商標庁との全ての業務を遂行するために、記名された発明者として、下記の弁護士及び/または弁理士を任命する。(氏名及び登録番号を記載すること)

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith (*list name and registration number)*

John H. Mion, Reg. No. 18,879; Thomas J. Macpeak, Reg. No. 19,292; Robert J. Seas, Jr., Reg. No. 21,092; Darryl Mexic, Reg. No. 23,063; Robert V. Sloan, Reg. No. 22,775; Peter D. Olexy, Reg. No. 24,513; J. Frank Osha, Reg. No. 24,625; Waddell A. Biggart, Reg. No. 24,861; Louis Gubinsky, Reg. No. 24,835; Neil B. Siegel, Reg. No. 25,200; David J. Cushing, Reg. No. 28,703; John R. Inge, Reg. No. 26,916; Joseph J. Ruch, Jr., Reg. No. 26,577; Sheldon I. Landsman, Reg. No. 25,430; Richard C. Turner, Reg. No. 29,710; Howard L. Bernstein, Reg. No. 25,665; Alan J. Kasper, Reg. No. 25,426; Kenneth J. Burchfiel, Reg. No. 31,333; Gordon Kit, Reg. No. 30,764; Susan J. Mack, Reg. No. 30,951; Frank L. Bernstein, Reg. No. 31,484; Mark Boland, Reg. No. 32,197; William H. Mandir, Reg. No. 32,156; Brian W. Hannon, Reg. No. 32,778; Abraham J. Rosner, Reg. No. 33,276; Bruce E. Kramer, Reg. No. 33,725; Paul F. Neils, Reg. No. 33,102; Brett S. Sylvester, Reg. No. 32,765; Robert M. Masters, Reg. No. 35,603; George F. Lehnigk, Reg. No. 36,359; John T. Callahan, Reg. No. 32,607; Steven M. Gruskin, Reg. No. 36,818; Peter A. McKenna, Reg. 38,551 and Edward F. Kenehan, Jr., Reg. 28,962

書類送付先：

Send Correspondence to:

SUGHRUE, MION, ZINN, MACPEAK & SEAS, PLLC
2100 Pennsylvania Avenue, N.W., Washington, D.C. 20037-3213

直通電話連絡先：(名称及び電話番号)

Direct Telephone Calls to: ***(name and telephone number)***

(202)293-7060

Full name of sole or first inventor 唯一又は第一発明者名	Nobuhiko TSUDA
Inventor's signature 発明者の署名	Date 日付
Residence 住所	Settsu-shi, OSAKA 566-8585 JAPAN
Citizenship 国籍	Japanese
Post office address 郵便の宛先	c/o Yodogawa-seisakusho, DAIKIN INDUSTRIES, LTD. 1-1, Nishihitotsuya, Settsu-shi, OSAKA 566-8585 JAPAN
Full name of second joint inventor, if any 第二共同発明者名(該当する場合)	Takahiro KITAHARA
Second inventor's signature 第二発明者の署名	Date 日付
Residence 住所	Settsu-shi, OSAKA 566-8585 JAPAN
Citizenship 国籍	Japanese
Post office address 郵便の宛先	c/o Yodogawa-seisakusho, DAIKIN INDUSTRIES, LTD. 1-1, Nishihitotsuya, Settsu-shi, OSAKA 566-8585 JAPAN

(第三以降の共同発明者についても同様に記載し、署名をすること。)

(Supply similar information and signature for third and subsequent joint inventors.)

Japanese Language Declaration
(日本語宣言書)

Full name of third joint inventor, if any 第三共同発明者名（該当する場合）	Ryuji IWAKIRI (deceased)	
Third inventor's signature 第三発明者の署名	MIYUKI IWAKIRI by Miyuki Iwakiri, as Legal representative of Ryuji Iwakiri, Deceased	Date 日付 March 27, 2001
Residence 住所	Residence of Miyuki Iwakiri: Minoo-shi, OSAKA 562-0005 JAPAN Residence of Ryuji Iwakiri (deceased): Settsu-shi, OSAKA 566-8585 JAPAN	
Citizenship 国籍	Citizenship of Miyuki Iwakiri: Japanese Citizenship of Ryuji Iwakiri (deceased): Japanese	
Post office address 郵便の宛先	Post office address of Miyuki Iwakiri: 11-31-102, Niina 4-chome, Minoo-shi, OSAKA 562-0005 JAPAN Post office address of Ryuji Iwakiri (deceased): c/o Yodogawa-seisakusho, DAIKIN INDUSTRIES, LTD. 1-1, Nishihitotsuya, Settsu-shi, OSAKA 566-8585 JAPAN	
Full name of fourth joint inventor, if any 第四共同発明者名（該当する場合）	Masaru NAGATO	
Fourth inventor's signature 第四発明者の署名		Date 日付
Residence 住所	Settsu-shi, OSAKA 566-8585 JAPAN	
Citizenship 国籍	Japanese	
Post office address 郵便の宛先	c/o Yodogawa-seisakusho, DAIKIN INDUSTRIES, LTD. 1-1, Nishihitotsuya, Settsu-shi, OSAKA 566-8585 JAPAN	
Full name of fifth joint inventor, if any 第五共同発明者名（該当する場合）	Ryoichi FUKAGAWA	
Fifth inventor's signature 第五発明者の署名		Date 日付
Residence 住所	Settsu-shi, OSAKA 566-8585 JAPAN	
Citizenship 国籍	Japanese	
Post office address 郵便の宛先	c/o Yodogawa-seisakusho, DAIKIN INDUSTRIES, LTD. 1-1, Nishihitotsuya, Settsu-shi, OSAKA 566-8585 JAPAN	
Full name of sixth joint inventor, if any 第六共同発明者名（該当する場合）		
Sixth inventor's signature 第六発明者の署名		Date 日付
Residence 住所		
Citizenship 国籍		
Post office address 郵便の宛先		

Japanese Language Declaration

日本語宣言

私は、米国法典第３５編１１９条(a)-(d)項又は３６５条(b)項に基き下記の、米国以外の国の少なくとも一ヵ国を指定している特許協力条約３６５(a)項に基ずく国際出願、又は外国での特許出願もしくは発明者証の出願についての外国優先権をここに主張するとともに、優先権を主張している、本出願の前に出願された特許または発明者証の外国出願を以下に、枠内をマークすることで、示しています。

I hereby claim foreign priority under Title 35, United States Code, § 119(a)-(d) or § 365 (b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

Prior foreign application(s) 外国での先行出願 (Number) (番号)	(Country) (国名)	(Day/Month/Year Filed) (出願年月日)	Priority Not Claimed 優先権主張なし
122384/1998	Japan	1/5/1998	☐
			☐
			☐

私は、第３５編米国法典１１９条(e)項に基いて下記の米国特許出願規定に記載された権利をここに主張いたします。

I hereby claim the benefit under Title 35, United States Code, § 119(e) of any United States provisional application(s) listed below.

(Application No.) (出願番号)	(Filing Date) (出願日)	(Application No.) (出願番号)	(Filing Date) (出願日)

私は、下記の米国法典第３５編１２０条に基いて下記の米国特許出願に記載された権利、又は米国を指定している特許協力条約３６５条(c)に基ずく権利をここに主張します。また、本出願の各請求範囲の内容が米国法典第３５編１１２条第１項又は特許協力条約で規定された方法で先行する米国特許出願に開示されていない限り、その先行米国出願書提出日以降で本出願書の日本国内または特許協力条約国際提出日までの期間中に入手された、連邦規則法典第３７編１条５６項で定義された特許資格の有無に関する重要な情報について開示義務があることを認識しています。

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s), or § 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

(Application No.) (出願番号)	(Filing Date) (出願日)	(Status)(patented, pending, abandoned) (現況：特許許可済、係属中、放棄済)

私は、私自身の知識に基ずいて本宣言書中で私が行なう表明が真実であり、かつ私の入手した情報と私の信じるところに基ずく表明が全て真実であると信じていること、さらに故意になされた虚偽の表明及びそれと同等の行為は米国法典第１８編第１００１条に基ずき、罰金または拘禁、もしくはその両方により処罰されること、そしてそのような故意による虚偽の声明を行なえば、出願した、又は既に許可された特許の有効性が失われることを認識し、よってここに下記のごとく宣誓を致します。

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Japanese Language Declaration

日本語宣言

委任状： 私は下記の発明者として、本出願に関する一切の手続きを米特許商標局に対して遂行する弁理士または代理人として、下記の者を指名いたします。（弁護士、または代理人の氏名及び登録番号を明記のこと）

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith: *(list name and registration number)*

John H. Mion, Reg. No. 18,879; Thomas J. Macpeak, Reg. No. 19,292; Robert J. Seas, Jr., Reg. No. 21,092; Darryl Mexic, Reg. No. 23,063; Robert V. Sloan, Reg. No. 22,775; Peter D. Olexy, Reg. No. 24,513; J. Frank Osha, Reg. No. 24,625; Waddell A. Biggart, Reg. No. 24,861; Louis Gubinsky, Reg. No. 24,835; Neil B. Siegel, Reg. No. 25,200; David J. Cushing, Reg. No. 28,703; John R. Inge, Reg. No. 26,916; Joseph J. Ruch, Jr., Reg. No. 26,577; Sheldon I. Landsman, Reg. No. 25,430; Richard C. Turner, Reg. No. 29,710; Howard L. Bernstein, Reg. No. 25,665; Alan J. Kasper, Reg. No. 25,426; Kenneth J. Burchfiel, Reg. No. 31,333; Gordon Kit, Reg. No. 30,764; Susan J. Mack, Reg. No. 30,951; Frank L. Bernstein, Reg. No. 31,484; Mark Boland, Reg. No. 32,197; William H. Mandir, Reg. No. 32,156; Brian W. Hannon, Reg. No. 32,778; Abraham J. Rosner, Reg. No. 33,276; Bruce E. Kramer, Reg. No. 33,725; Paul F. Neils, Reg. No. 33,102; Brett S. Sylvester, Reg. No. 32,765; Robert M. Masters, Reg. No. 35,603, George F. Lehnigk, Reg. No. 36,359, John T. Callahan, Reg. No. 32,607 and Steven M. Gruskin, Reg. No. 36,818

32

書類送付先

Send Correspondence to:
SUGHRUE, MION, ZINN, MACPEAK & SEAS, PLLC
2100 Pennsylvania Avenue, N.W. Suite 800
Washington, D.C. 20037-3213

直接電話連絡先： （名前及び電話番号）

Direct Telephone Calls to:

(202)293-7060

唯一または第一発明者名 1-00		Full name of sole or first inventor Nobuhiko TSUDA
発明者の署名 日付		Inventor's signature Nobuhiko Tsuda Dec. Date 12, 2000
住所		Residence Settsu-shi, OSAKA 566-8585 JAPAN JPX
国籍		Citizenship Japanese
私書箱		Post Office Address c/o Yodogawa-seisakusho, DAIKIN INDUSTRIES, LTD., 1-1, Nishihitotsuya, Settsu-shi, OSAKA 566-8585 JAPAN
第二共同発明者 2-00		Full name of second joint inventor, if any Takahiro KITAHARA
第二共同発明者 日付		Second inventor's signature Takahiro Kitahara Dec. Date 12, 2000
住所		Residence Settsu-shi, OSAKA 566-8585 JAPAN JPX
国籍		Citizenship Japanese
私書箱		Post Office Address c/o Yodogawa-seisakusho, DAIKIN INDUSTRIES, LTD., 1-1, Nishihitotsuya, Settsu-shi, OSAKA 566-8585 JAPAN

（第三以降の共同発明者についても同様に記載し、署名をすること）

(Supply similar information and signature for third and subsequent joint inventors.)

Japanese Language Declaration

日本語宣言書

第三の共同発明者の氏名（該当する場合）	Full name of third joint inventor, if any Ryuji IWAKIRI (deceased)
同第三発明者の署名　日付	Third inventor's signature　Date Miyuki Iwakiri Dec. 12, 2000 by Miyuki Iwakiri, as Legal representative of Ryuji Iwakiri, Deceased
住所	Residence Minoo-shi, OSAKA 562-0005 JAPAN JPX
国籍	Citizenship Japanese
郵便の宛先	Post office address 11-31-102, Niina 4-chome, Minoo-shi, OSAKA 562-0005 JAPAN
第四の共同発明者の氏名（該当する場合）	
同第四発明者の署名　日付	Full name of fourth joint inventor, if any Masaru NAGATO
住所	Fourth inventor's signature　Date Masaru Nagato Dec. 12, 2000
国籍	Residence ~~Settsu-shi, OSAKA~~ 566-8585 JAPAN JPX
郵便の宛先	Citizenship Japanese
	Post office address c/o Yodogawa-seisakusho, DAIKIN INDUSTRIES, LTD., 1-1, Nishihitotsuya, Settsu-shi, OSAKA 566-8585 JAPAN
第五の共同発明者の氏名（該当する場合）	
同第五発明者の署名　日付	Full name of fifth joint inventor, if any Ryoichi FUKAGAWA
住所	Fifth inventor's signature　Date Ryoichi Fukagawa Dec. 12, 2000
国籍	Residence Settsu-shi, OSAKA 566-8585 JAPAN JPX
郵便の宛先	Citizenship Japanese
	Post office address c/o Yodogawa-seisakusho, DAIKIN INDUSTRIES, LTD., 1-1, Nishihitotsuya, Settsu-shi, OSAKA 566-8585 JAPAN
第六の共同発明者の氏名（該当する場合）	
同第六発明者の署名　日付	Full name of sixth joint inventor, if any
住所	Sixth inventor's signature　Date
国籍	Residence
郵便の宛先	Citizenship
	Post office address